

## PATENT COOPERATION TREATY

**PCT****NOTIFICATION OF ELECTION**

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Assistant Commissioner for Patents  
United States Patent and Trademark  
Office  
Box PCT  
Washington, D.C.20231  
ETATS-UNIS D'AMERIQUE

in its capacity as elected Office

<b>Date of mailing (day/month/year)</b> 29 March 2000 (29.03.00)	<b>Applicant's or agent's file reference</b> PBA/DO88499PWO
<b>International application No.</b> PCT/GB99/02420	<b>Priority date (day/month/year)</b> 06 August 1998 (06.08.98)
<b>International filing date (day/month/year)</b> 06 August 1999 (06.08.99)	
<b>Applicant</b> SALISBURY, Richard, James	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:  
03 March 2000 (03.03.00)

☐ in a notice effecting later election filed with the International Bureau on:  
\_\_\_\_\_

2. The election ☒ was  
☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32<sup>1</sup> applies, within the time limit under Rule 32.2(b).

<b>The International Bureau of WIPO</b> 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No.: (41-22) 740.14.35	<b>Authorized officer</b> Juan Cruz Telephone No.: (41-22) 338.83.38
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## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference PBA/DO88449PWO	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/GB99/02420	International filing date (day/month/year) 06/08/1999	Priority date (day/month/year) 06/08/1998
International Patent Classification (IPC) or national classification and IPC B27N3/00		
Applicant UNIVERSITY OF WALES, BANGOR et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 5 sheets, including this cover sheet.

- ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand  03/03/2000	Date of completion of this report  23.11.2000
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer  Baath, C  Telephone No. +49 89 2399 2959 

# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB99/02420

## I. Basis of the report

1. This report has been drawn on the basis of *(substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments (Rules 70.16 and 70.17).):*  
**Description, pages:**

1-12 as originally filed

### **Claims, No.:**

1-24 as originally filed

### **Drawings, sheets:**

1/9-9/9 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:

# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB99/02420

☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

## V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

### 1. Statement

Novelty (N)	Yes: Claims	
	No: Claims	1-3
Inventive step (IS)	Yes: Claims	4, 6-17, 21-24
	No: Claims	
Industrial applicability (IA)	Yes: Claims	1-24
	No: Claims	

2. Citations and explanations  
**see separate sheet**

## VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:  
**see separate sheet**

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/GB99/02420

**V Novelty and inventive step**

**1. Claim 1**

JP abstract 15 32 72 discloses

a method of forming products from lignocellulosic material in which said material is subjected to a binding operation using a phenol formaldehyde resin which is cured during said operation wherein at least one of maleic acid and maleic anhydride is added during the formation of such products so as to be in admixture with the resin when it is cured.

The requirement of Art. 33 (2) is not met.

**2. Claims 2 and 3**

The JP abstract describes 5-15 parts of maleic acid or maleic anhydride in the resin, which corresponds to or is in the same range as the claimed values of 5-55 % or 15-40 %.

The requirement of Art.33 (2) is not satisfied.

**3. Further analysis**

Claim 4

For the purpose of avoiding a non-favourable influence of the maleic agent onto the viscosity of the phenol formaldehyde resin mixture (see page 3, par. 2) the concept of

applying the maleic acid or maleic anhydride to the lignocellulosic material separately of the application of the phenol formaldehyde resin remains without example in the prior art as currently available.

A new independent claim 1 which comprises the features of current claims 1-4 and acknowledges in a preamble the prior art according to the JP abstract, i. e. the features of current claims 1-3, Rule 6.3 a), b) would have satisfied the requirement of Art. 33 (3).

Current claims 6-17 and 21-24 could have been maintained as dependent on such a claim 1.

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/GB99/02420

**VIII Clarity**

1. Current claim 5 contradicts the teaching of current claim 4. Although disclosed in the current application, the conditions under which such a mixing with the phenol resin can be tolerated without the cited adverse effect of increasing viscosity has not been discussed. The requirements of Art. 6 and Rule 5.1 iii) are not satisfied. Moreover, mixing the maleic agent with the phenol formaldehyde agent is known from the said JP abstract, Art. 33 (2) is not met.
2. Claim 18 appears to be a mere repetition of what is claimed in claim 1. Claim 18 contradicts the requirement of conciseness, Art. 6. The same applies to claims 19 and 20.

## PCT

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>PBA/D088499PWO</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/GB 99/ 02420</b>	International filing date (day/month/year) <b>06/08/1999</b>	(Earliest) Priority Date (day/month/year) <b>06/08/1998</b>
Applicant <b>UNIVERSITY OF WALES, BANGOR et al.</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 3 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

**1. Basis of the report**

- a. With regard to the language, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

- b. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international search was carried out on the basis of the sequence listing:

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

2. ☐ Certain claims were found unsearchable (See Box I).

3. ☐ Unity of invention is lacking (see Box II).

**4. With regard to the title,**

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

**5. With regard to the abstract,**

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

**6. The figure of the drawings to be published with the abstract is Figure No.**

☐ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

☒ None of the figures.





## INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 99/02420

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 97 32074 A (WINDSOR TECHNOLOGIES LIMITED ; SYMONS MICHAEL WINDSOR (ZA)) 4 September 1997 (1997-09-04) page 5, line 13 - line 28 page 6, line 16 - line 22 page 8, line 1 - line 10 page 10, line 14 - line 20	1
A	US 5 520 777 A (SHISKO WALTER S) 28 May 1996 (1996-05-28) cited in the application abstract; claim 1	1

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/GB 99/02420

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
JP 47018996	B		NONE	
US 3968294	A	06-07-1976	US 4044087 A	23-08-1977
WO 9732074	A	04-09-1997	AU 706508 B	17-06-1999
			AU 1803797 A	16-09-1997
			EP 0880619 A	02-12-1998
US 5520777	A	28-05-1996	AU 1803195 A	11-09-1995
			CA 2184268 A	31-08-1995
			WO 9523055 A	31-08-1995

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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 7 : <b>B27N 3/00, C08L 97/02</b>	<b>A1</b>	(11) International Publication Number: <b>WO 00/07786</b> (43) International Publication Date: 17 February 2000 (17.02.00)
<p>(21) International Application Number: <b>PCT/GB99/02420</b></p> <p>(22) International Filing Date: <b>6 August 1999 (06.08.99)</b></p> <p>(30) Priority Data: <b>9817094.7</b> <b>6 August 1998 (06.08.98)</b> <b>GB</b></p> <p>(71) Applicant (for all designated States except US): <b>UNIVERSITY OF WALES, BANGOR [GB/GB]; The Biocomposites Centre, University of Wales, Bangor, Gwynedd LL57 2UW (GB).</b></p> <p>(72) Inventor; and (75) Inventor/Applicant (for US only): <b>SALISBURY, Richard, James [GB/GB]; Berth LWYD, Tyn Y Gongl, Anglesey LL74 8NS (GB).</b></p> <p>(74) Agent: <b>ATKINSON, Peter, Birch; Marks &amp; Clerk, Sussex House, 83-85 Mosley Street, Manchester M2 3LG (GB).</b></p>	<p>(81) Designated States: <b>AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).</b></p> <p><b>Published</b> <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i></p>	
<p>(54) Title: <b>BONDING LIGNOCELLULOSIC MATERIALS</b></p> <p>(57) Abstract</p> <p>The invention relates to a method of forming products from lignocellulosic material in which said material is subjected to a binding operation using a phenol formaldehyde resin which is cured during said operation wherein at least one of maleic acid and maleic anhydride is added during the formation of such products so as to be in admixture with the resin when it is cured.</p>		

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BONDING LIGNOCELLULOSIC MATERIALS

The present invention relates to a method of forming products from lignocellulosic material in which the lignocellulosic material is subjected to a bonding operation using a phenol formaldehyde resin which is cured during said operation. The invention relates particularly (but not exclusively) to such methods in which lignocellulosic materials are bonded together, e.g. the production of phenolic resin bonded particle board and the production of plywood and other glued wood products.

The manufacture of particle boards involves the steps of forming a mixture of wood particles, a phenolic resin, and other additives and forming the mixture into what is known as a mattress. This mattress is then pressed into its final shape while heat is supplied to cure the resin. The time required to cure the resin so as to achieve satisfactory products is one of the factors determining the cost of the finished product as it governs the throughput that can be achieved. A satisfactory cure is needed to ensure that the final properties of the board are adequate for any particular end use. Such properties include water resistance (which is measured by examining the swelling of finished boards after soaking in water, and internal bond strength) since water can cause products made in this way to deteriorate resulting in a loss of bond strength with consequential swelling of the board and, in severe cases disintegration of the board. It is therefore desirable to improve the ability of such products to resist attack by water so as to obtain products with high dimensional stability.

GB-A-2 136 004 discloses the pre-treatment of wood particles with dibasic anhydrides to produce an esterified woody material which is used to form a moulding by mixing with an epoxy resin and effecting curing. The esterification reaction is carried out in the presence of a catalyst and, as exemplified, the reaction times vary from 7.5 minutes to 15 hours, the longest reaction times being used where the wood particles are chips of the kind used for producing particle boards. This prior specification includes a comparative example in which a particle board is made using a phenolic resin, but there is no suggestion of using the esterified wood in the manufacture of particle boards.

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US-A-5 520 777 discloses a method of making fibreboard which includes forming a condensate of maleic anhydride and glycerol and impregnating wood chips with this condensate before the chips are transferred to a digester where, during steaming at an elevated pressure, an esterification reaction takes place between the components of the wood and the condensate. After digestion, the treated material is refined i.e. converted to fibre form under pressure. US-A-4 961 795 acknowledges at column 1, lines 35 to 45 that acids have been added to resole resins to accelerate the cure of phenol formaldehyde resins, and the prior art cited against this specification includes several specifications in which acids are added to phenol formaldehyde resins, either to control the rate of curing, or to increase the shelf life of resole resins whose initial formation involved manufacture at relatively high alkaline pH, as the higher the pH the shorter the shelf life of the resin. This means that if resins are e.g. manufactured at pH 11, the pH will be reduced to say 7 by the addition of e.g. sulphuric or oxalic acid before the resin is sold for use. US-A-4 001 186 describes such a process for stabilising a resin. US-A-4 961 795 describes the use of curing agents for phenol-formaldehyde which have an ester functional group selected from the group consisting of lactones, organic carbonates, carboxylic acid esters or mixtures thereof. These curing agents are added to the resin prior to its addition as a binder, and in order to avoid unacceptable increases in the viscosity of the resin so reducing the pot life of the resin as it becomes too viscous to handle a further additive in the form of an aliphatic alcohol is added to moderate the effect of the curing agent. There is no teaching in the specification concerning the pH at which curing takes place, but where possible to deduce this from the examples, it appears curing takes place at alkaline or neutral conditions.

According to the present invention there is provided a method of forming products from lignocellulosic material in which said material is subjected to a bonding operation using a phenol formaldehyde resin which is cured during said operation wherein at least one of maleic acid and maleic anhydride is added during the formation of such products so as to be in admixture with the resin when it is cured.

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We have found that the inclusion of maleic anhydride and/or maleic acid in the phenol formaldehyde resin to be cured can be used either to achieve cost savings by reducing the quantity of resin required to achieve a product with adequate performance, or to enhance the performance that can be achieved with a particular quantity of resin and particular manufacturing conditions.

In the case where the resin is a solid (powder), the maleic anhydride and/or maleic acid may be incorporated in the resin to be cured by admixture with the resin prior to its application to the lignocellulosic material to be bonded. In the case where the resin is a liquid, the maleic anhydride and/or maleic acid may be incorporated into the liquid prior to application to lignocellulosic material. However, in this case, incorporation of the maleic acid and/or maleic anhydride into the liquid resin may affect the viscosity thereof and this may not be desirable, e.g. where it is desirable to spray the mixture of resin and maleic additive onto the lignocellulosic material. Alternatively, the maleic anhydride and/or maleic acid may be applied to the lignocellulosic material separately to application of the resole resin.

Therefore in contrast with the process disclosed in US-A-5 520 777, we have found that it is not necessary to go to the expense of converting the maleic anhydride into a condensate with glycerol. We can also avoid the need to impregnate the lignocellulose component since it is possible to merely apply the maleic anhydride or maleic acid either to the lignocellulose surface or admix it with the phenol formaldehyde resin. Additionally, the present invention avoids the extreme conditions required to esterify the lignocellulose.

If maleic acid is used as the additive for the phenol formaldehyde resin then it is possible that some or all of the maleic acid takes part in the curing process through conversion to the anhydride. The ability of maleic anhydride to become incorporated into the resin matrix is probably due to the electrophilic nature of the maleic anhydride molecule under the conditions used in forming a cured phenolic resin and the likelihood that it provides cross-links in the cured resin. We have found that other acidic anhydrides such as phthalic and succinic anhydride do not produce the

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same benefit, possibly because they do not possess a suitable combination of acidic and electrophilic properties when curing a phenol formaldehyde resin. Strong acids, such as sulphuric acid, while capable of producing an improved product in terms of bond strength and water resistance, cannot be used because of the possibility of causing corrosion of the manufacturing plant, and in addition cause discoloration of the resin which can be seen in the finished product, which is unacceptable from a marketing point of view. Additionally, such strong acids can cause degradation of the board. Maleic acid and anhydride, though weaker acids, unexpectedly give a product equivalent in performance without the disadvantages arising from the use of strong acids.

Whilst the invention includes the use of mixtures of maleic anhydride and maleic acid there is however no advantage in using such a mixture as it simply means that two raw materials have to be utilised rather than choosing one. Moreover, for reasons of cost, maleic anhydride is preferred to maleic acid as the additive for use in the invention.

The phenol formaldehyde resin for use in the invention may be a novolac resin, in which case the maleic anhydride and/or maleic acid may substitute for at least a part of the curing agent (generally a compound capable of yielding formaldehyde) used for the cure of such resins. Furthermore, the maleic additive may substitute for at least part of the acidic curing agent normally used with such resins.

It is however more preferred that the phenol formaldehyde resin used in the method of the invention is a resole resin which may be an oligomer which is obtained by reaction of formaldehyde and phenol in the presence of an alkali, the oligomer being polymerisable when heated during pressing to form the cured resin which provides the bond.

It is preferred that the resole resin has a pH of at most 11.5, more preferably at most 11.0, and even more preferably at most 10.5. A particularly suitable pH is about 10.



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An example of resole resin which may be used in the method of the invention is that sold by Neste Chemicals Ltd under the designation BD937.

The invention is applicable particularly in the bonding together of lignocellulosic material (e.g. in the manufacture of glued wood products, e.g. plywood) but may also be applied to the bonding of lignocellulosic materials to other substrates.

The invention finds particularly utility in the manufacture of particle board (for which the lignocellulosic material (e.g. wood or other plant residues) to be bonded together may be in the form of fibre, chips, shavings and/or flakes) and will be further described with reference to such products.

The invention therefore also includes in a process for manufacturing phenolic bonded wood particle board in which a mixture of lignocellulose particles and a phenolic resin are formed into a cured board under the action of heat and pressure, the improvement of providing in the mixture prior to pressing to form the cured board, at least one of maleic anhydride and maleic acid.

We have found that maleic anhydride and maleic acid can assist in obtaining a larger throughput of boards, because of a faster cure, and also gives an improvement in internal bond strength and water resistance over boards made under comparable conditions in the absence of the maleic anhydride acid additive. Moreover, by using maleic anhydride as the additive, the cost of the product may also be reduced since maleic anhydride costs less than the phenolic resin for which it may in part be substituted.

The quantity of maleic anhydride or maleic acid added is preferably in the range 5-55% by weight of the total weight of the resin and maleic anhydride and/or maleic acid, and more preferably 15-40% by weight on the same basis.

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The combined amount of resin and maleic anhydride or acid used will vary according to the requirements of the product but usually ranges from 2% to 15% by weight of dry wood. Measurements are based in dry wood content but the wood chips used can contain as much as 14% by weight of water, and do not necessarily need to be dried before use. In this respect, we have found that maleic anhydride and maleic acid have the advantage of reducing sensitivity of the process (of producing particle board) to moisture and limit the effect of excess water on cure.

Maleic anhydride may be added as a solid or a liquid to the lignocellulose particles at the same time as the resin, or incorporated into the lignocellulose particle mix separately to the resin. It is also possible to spray molten maleic anhydride onto the lignocellulosic particles. Similar techniques may also be used in respect of maleic acid. The invention also includes dissolving maleic anhydride in water, thus forming a maleic acid solution and adding the maleic anhydride in this way to the system. It is possible that some or all of the maleic acid may react in the system in the anhydride form through decomposing back to the anhydride during the application of heat and pressure.

In forming phenolic resin lignocellulose particle boards, it is common to incorporate a wax emulsion into the mix. Maleic anhydride or maleic acid may be admixed into the wax emulsion prior to its addition to the lignocellulose particle mix.

A suitable wax was emulsion of a microcrystalline wax is that sold under the trade name Mobilcer 538 by Mobil Ltd. We have found that a mixture of maleic acid and wax emulsion is made more stable by the further addition of a polybutene emulsion such as that sold by BP Chemicals under the trade name Hyvis polybutene.

Wax is sometimes added in the manufacture of particle board in a molten form, and in this case is usually sprayed onto the lignocellulose particles. In such a case, molten maleic anhydride may be sprayed on to the lignocellulose particles before, simultaneously with (but separately), admixed with, or after spraying with molten wax. Molten maleic anhydride may also be used in the absence of wax. It can

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conveniently be sprayed onto e.g. lignocellulose chips during the final stage of a drying process. Similar techniques may also be employed with maleic acid.

A well known commercial form of particle board is oriented strand board (OSB). OSB is made from wood strands produced by slicing and milling wood. The strands will typically have dimensions of the order 50mm x 10mm x 2mm. The strands after drying are sprayed with hot wax and mixed with powdered phenolic resin. The strands are then formed into a mattress which is typically pressed at 200°C to cure the resin. The pressing time is measured in so many seconds per mm thickness of the mattress, and is typically chosen from the range 10-20 seconds per mm. The present invention could be applied to such a commercial process by adding maleic acid solution to the wood in the dryer, or spraying molten maleic anhydride along with the hot wax. The molten wax can also be replaced by a wax emulsion to which a solution of maleic acid has been added.

Although the invention has been described with particular reference to the production of particle board, it will be appreciated that the invention is applicable to other products obtained by bonding lignocellulosic materials. Examples of such products in which lignocellulosic materials are bonded together include plywood and other glued wood products. The invention is also applicable to the bonding of lignocellulosic materials to other substrates and may thus be applied, for example, to the production laminates from lignocellulose and resin impregnated paper, e.g. for the purpose of producing a decorative laminate.

The invention is further illustrated by the following non-limiting Examples and accompanying Figs 1 to 9 which illustrates the results of the Examples.

In all of the Examples the resole resin used was a commercially available resole resin sold by Neste Chemicals Ltd under the designation BD937. The boards were pressed at 200°C in a 150mm die to 12mm stops with a pre-heated punch. The test samples were conditioned according to the method BS5669 before being tested for internal bond strength and thickness swell after 24 hours soaking in water at 20°C,

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and retained internal bond after drying at 60°C and re-conditioning according to the method of BS5669.

### Example 1

A series of boards was made using softwood chips with a moisture content of 4%, resin powder, various acids, and a mixture consisting of 80% by weight of Mobilcer 538 and 20% polybutene emulsion. The quantity wax mixture was 1.6% of the dry weight of wood. The various acids were mixed with the wax mixture before the wax/acid mixture was added to the wood. The quantity of maleic acid was used in the proportion of one part acid to three parts resin, dry weights. The other acids were added in sufficient quantity to maintain an equivalent molar ratio of acid to resin to that achieved by the maleic acid example. After mixing in the wax, sufficient resin was added to make the total dry weight of acid and dry weight of resin up to 5% of the dry weight of wood. Boards were obtained by pressing for three minutes.

The internal bonds and thickness swells of the boards were measured and the results are shown in Fig 1. The effect of the acids used on the colour of the cured resin were also observed with the following results:

Acidic material	pKa	resin colour
Sulphuric acid	-9	Black
p-toluene sulphonic acid	-6.5	Black
trichloroacetic acid	0.7	dark red
dichloroacetic acid	1.48	light yellow
maleic acid	1.83	light yellow

It can be seen from Fig 1, that (with the exception of maleic acid) the acids having a pKa value greater than 0 tended to give inferior internal bond strength and higher thickness swells than sulphuric acid and p-toluene sulphonic acid. However somewhat unexpectedly, maleic acid gave internal bond strengths and thickness

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swelling values commensurate with those obtained using sulphuric acid and p-toluene sulphonic acid but had the advantage of providing a resin of light yellow colour as compared to the black cured resin obtained using sulphuric acid p-toluene sulphonic acid.

### Example 2

A series of boards were made to demonstrate the improved properties obtained by the addition of maleic acid. The wood was a soft wood shred with a moisture content of about 6 per cent. The wood shred was wetted with 2% by weight (based on the dry weight of wood) of Molibcer 538, then 5% by weight of resin (again, dry wood basis) was stirred in.

Boards were made with either no maleic acid or with 25% or 40% of the resin replaced by maleic acid. The maleic acid was added to the dried wood shred as a 30% aqueous solution (calculated as weight of anhydride to weight of solution) made by dissolving maleic anhydride in water. The acid solution was added before the wax dispersion. The boards were pressed for 2½, 3, 3½, 4, and 5 minutes.

Fig 2 shows the effect of replacing 40% of the resin with maleic acid on internal bond strength (IBS) obtained after pressing for the times and temperature specified. Fig 3 shows (again for 40% resin replacement) the IBS after soaking the series of boards in water for 24 hours at 20°C. Fig 4 shows the degree of thickness swell after the same treatment (for boards obtained by replacing 40% by resin).

Figs 5 to 7 are similar to Figs 2 to 4 but showing the result obtained with 25% resin replacement.

The results of these experiments clearly show the benefit of the addition of maleic acid achieving the development of satisfactory IBS, both initially and after soaking, as well as swell resistance. One way of expressing the improvements achieved is in the terms of the acceleration in the curing time in the reduction in time

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required to achieve a particular strength, both initially and after soaking, to in the time to achieve a low level of thickness swell. It can be seen that with the addition of maleic acid both initial IBS development and IBS after cold soak is accelerated by about 4-6 sec/mm and swell resistance by about 5sec/mm.

### Example 3

Either powdered maleic anhydride or powdered maleic acid was mixed with powdered BD937 resin in the ratio of one part to three parts. 3g of Mobilcer 538 wax emulsion was stirred into 265g of wood chips with a moisture content of 10% for one minute. 12g of the mixture of resin and maleic anhydride was stirred in for a further minute. Control boards using 12g BD937 were made according to the same method. The boards were pressed for 3 minutes, 3½ minutes or 4 minutes. The boards made with maleic anhydride or maleic acid cured more quickly, had higher internal bond strength, swelled less in cold water, and had higher retained internal bond strength after soaking, as shown in Fig 8.

### Example 4

BD937 resin was made into a 50%, by weight, aqueous solution. 7.5% of the BD937 resin solution was mixed with wood chips, with a moisture content of 8.7%, for one minute and then 1.25%, by weight of dry wood, of maleic anhydride was mixed in for a further 1 minute. Control boards were made by mixing 10% of the BD937 resin solution, by weight of dry wood, with the wood chips. Boards were pressed for 3, 3½, 4, or 5 minutes. The boards made with maleic anhydride had higher internal bond strength, swelled less in cold water and had higher retained internal bond strength after soaking, as shown in Fig 9.

Examples 5 to 13 are provided to illustrate the various ways in which maleic acid and maleic anhydride may be added to improve the performance of resole resins. Boards were obtained by pressing for 2½ to 5 minutes and when tested gave results of the same order as this obtained in Examples 1 to 4.

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Example 5

Maleic anhydride was stirred into water to make a 30% w/w solution of maleic acid. Mobilcer 538 wax emulsion was mixed with Hyvis polybutene emulsion in the ratio 3:1. The maleic acid solution was mixed with the wax mixture in the ratio 10:3. 13g of the mixture was stirred into 250g of wood chips with a moisture content of 5.4% for 1 minute, followed by 9g powdered BD937 resin which was stirred in for 3 minutes.

Example 6

Maleic anhydride was stirred into water to make a 30% w/w solution of maleic acid. 10g of this mixture and 3g of Mobilcer 538 wax emulsion were stirred into 250g of wood chips with a moisture content of 5.4% for 1 minute, followed by 9g powdered BD937 resin which was stirred in for 3 minutes.

Example 7

Maleic anhydride was stirred into water to make a 30% w/w solution of maleic acid. 10g of this solution was sprayed onto 250g of wood chips with a moisture content of 11% while they were being stirred for 1 minute. 9g of powdered BD937 resin was stirred in for 1 minute.

Example 8

BD937 resin was made into a solution of 50% solids content. 18g of this solution was stirred into 250g of wood chips with a moisture content of 11% for one minute. 3g of powdered maleic anhydride was then stirred in for 1 minute.

Example 9

BD937 resin was made into a solution of 50% solids content. 18g of this solution and 3g of powdered maleic anhydride were stirred into 250g of wood chips with a moisture content of 11% for 2 minutes.

Example 10

BD937 resin was made into a solution of 50% solids content. 18g of this solution was stirred into 250g of woods chips with a moisture content of 11% for one minute. 3g of powdered maleic acid was then stirred in for one minute.

Example 11

BD937 resin was made into a solution of 50% solids content. 18g of this solution and 3g of powdered maleic acid were stirred into 250g of wood chips with a moisture content of 11% for 2 minutes.



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CLAIMS

1. A method of forming products from lignocellulosic material in which said material is subjected to a binding operation using a phenol formaldehyde resin which is cured during said operation wherein at least one of maleic acid and maleic anhydride is added during the formation of such products so as to be in admixture with the resin when it is cured.
2. A method as claimed in claim 1 wherein the amount of maleic anhydride and/or maleic acid is 5% to 55% by weight based on the total weight of the resin and the/or maleic acid.
3. A method as claimed in claim 2 wherein the amount of maleic anhydride and/or maleic acid is 15% to 40% by weight based on the total weight of the resin and/or maleic acid.
4. A method as claimed in any one of claims 1 to 3 wherein the maleic acid and/or maleic anhydride is applied to the lignocellulosic material separately of application of the resin.
5. A method as claimed in any one of claims 1 to 3 wherein the maleic acid and/or maleic anhydride is admixed with the resin separately to application of the admixture of the lignocellulosic material.
6. A method as claimed in any one of claims 1 to 5 wherein the maleic acid and/or maleic anhydride are admixed with a wax emulsion.
7. A method as claimed in claim 6 wherein the wax emulsion is rendered more stable by the inclusion of a polybutene emulsion.

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8. A method as claimed in any one of claims 1 to 7 which utilises maleic anhydride.
9. A method as claimed in any one of claims 1 to 8 wherein the phenol formaldehyde resin is a novolac resin.
10. A method as claimed in any one of claims 1 to 7 wherein the phenol formaldehyde is a resole resin.
11. A method as claimed in claim 10 wherein the resole resin has a pH of at most 11.5.
12. A method as claimed in claim 11 wherein the resole resin has a pH of at most 11.0.
13. A method as claimed in claim 12 wherein the resole resin has a pH of at most 10.5.
14. A method as claimed in claim 13 wherein the resole resin has a pH of about 10.
15. A method as claimed in any one of claims 1 to 14 wherein the method is applied to the bonding of lignocellulosic material to lignocellulosic material.
16. A method as claimed in claim 15 wherein the lignocellulosic material to be bonded is in the form of particles.
17. A method as claimed in claim 16 wherein the product is particle board.
18. In a method for manufacturing phenolic bonded lignocellulosic in which a mixture of lignocellulose particles and a phenolic resin are formed into a cured board under the action of heat and pressure, the improvement of providing in the mixture

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prior to pressing to form the cured board, at least one of maleic anhydride and maleic acid.

19. A method as claimed in any one of claims 15 to 18 wherein the amount of resin and maleic anhydride and/or maleic acid is 2% to 15% by weight of the dry lignocellulosic material.

20. A method as claimed in any one of claims 15 to 19 wherein the lignocellulosic material has a maximum water content of 14% by weight.

21. A method as claimed in any one of claims 16 to 20 wherein the product is orientated strand board.

22. A method as claimed in claim 15 wherein the product is plywood or other glued wood product.

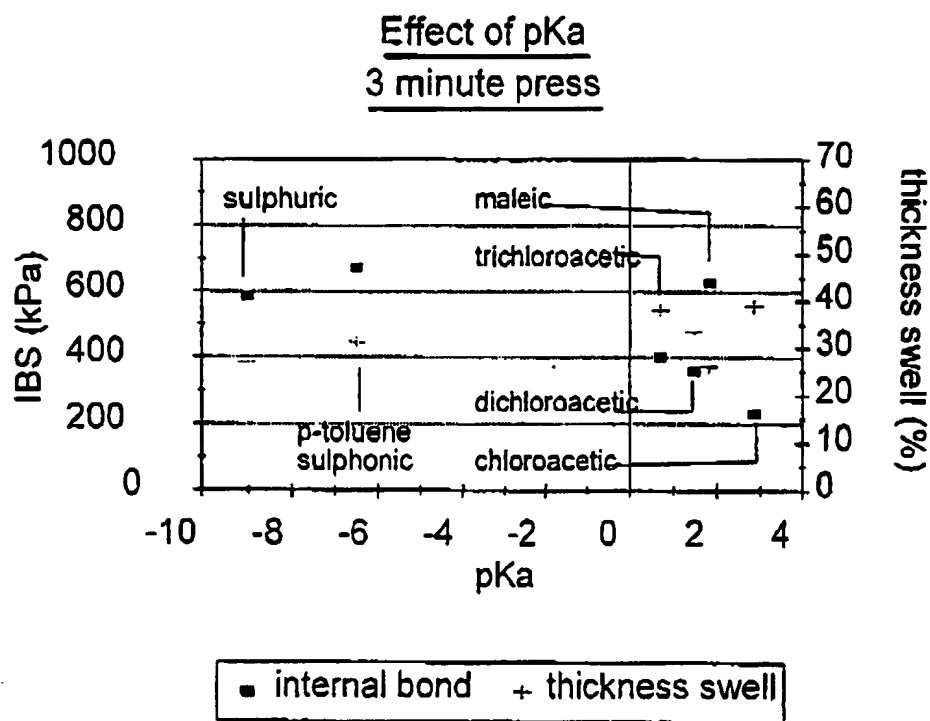
23. A method as claimed in any one of claims 1 to 14 applied to the bonding of lignocellulosic material to non-lignocellulosic material.

24. A method as claimed in claim 23 wherein the non-lignocellulosic material is a resin impregnated paper.

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Fig.1

Effect of replacing 40% of resin  
with maleic acid on IBS

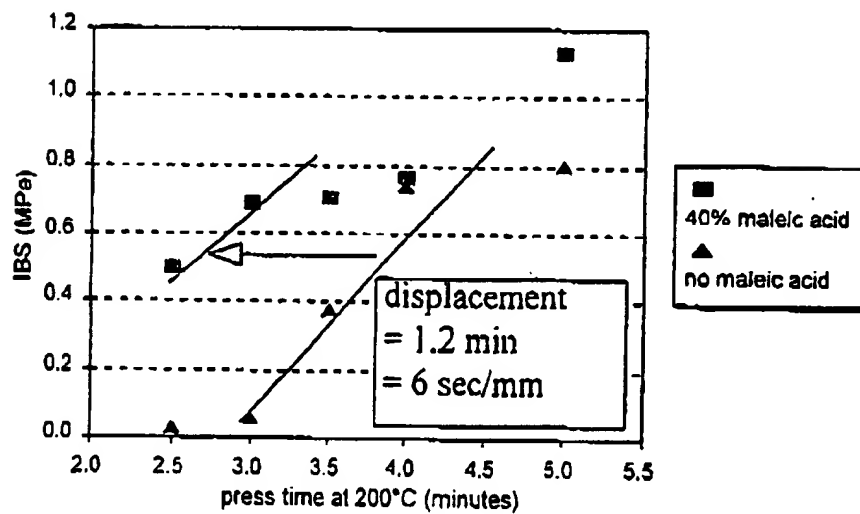


Fig.2

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Effect of replacing 40% of resin with  
maleic acid on IBS after cold soak

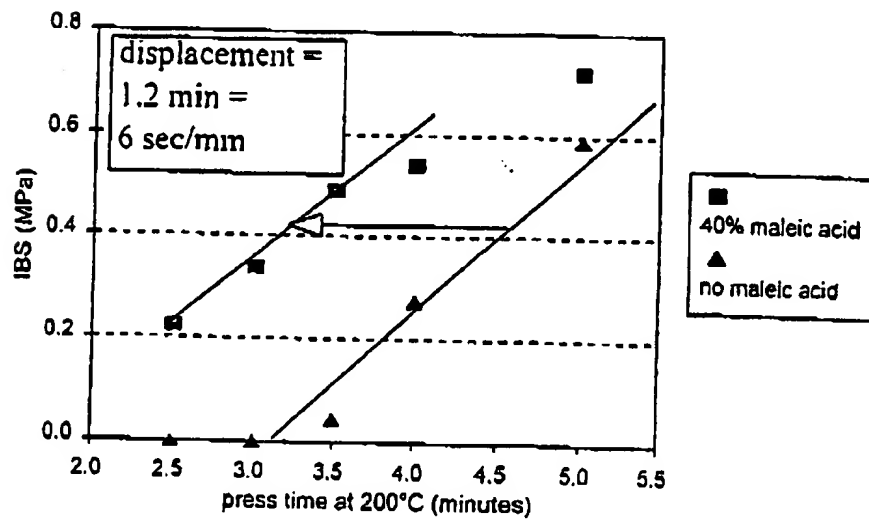


Fig.3

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Effect of replacing 40% of resin with  
maleic acid on thickness swell

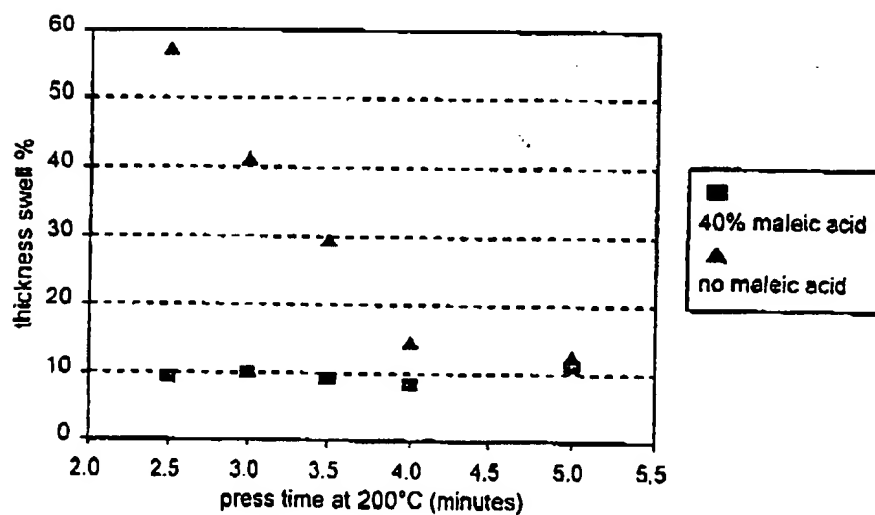


Fig4

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Effect of replacing 25% of resin  
with maleic acid on IBS

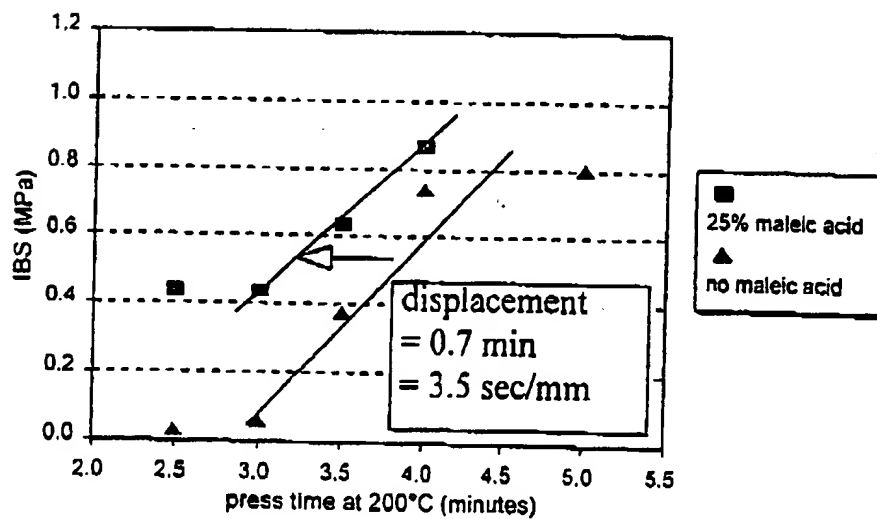


Fig.5



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Effect of replacing 25% of resin with  
maleic acid on IBS after cold soak

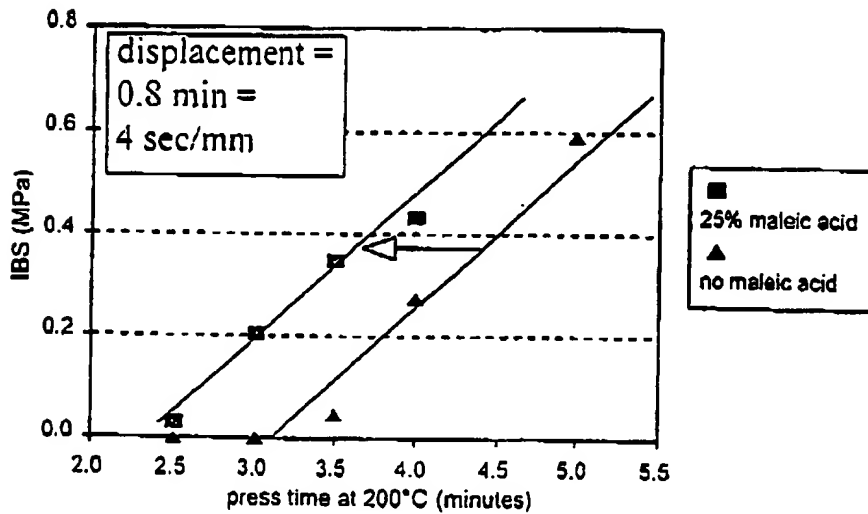


Fig.6

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Effect of replacing 25% of resin with  
maleic acid on thickness swell

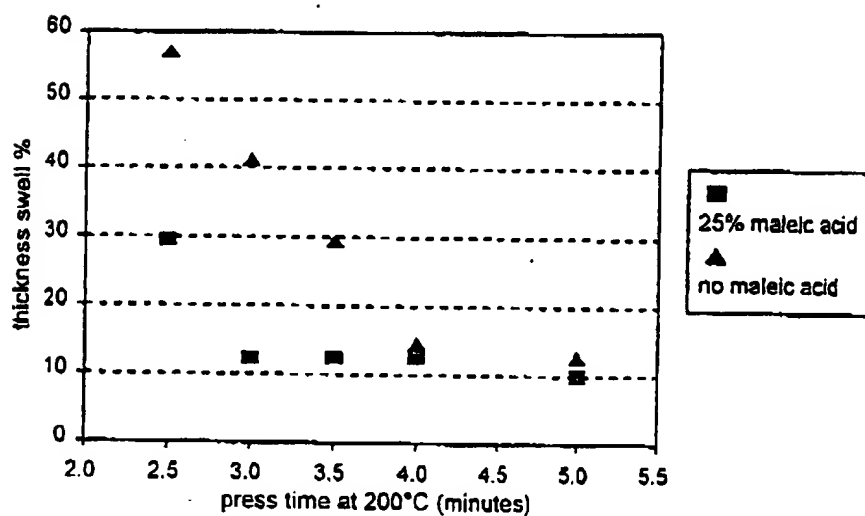


Fig.7

### Retained Internal Bond after soaking

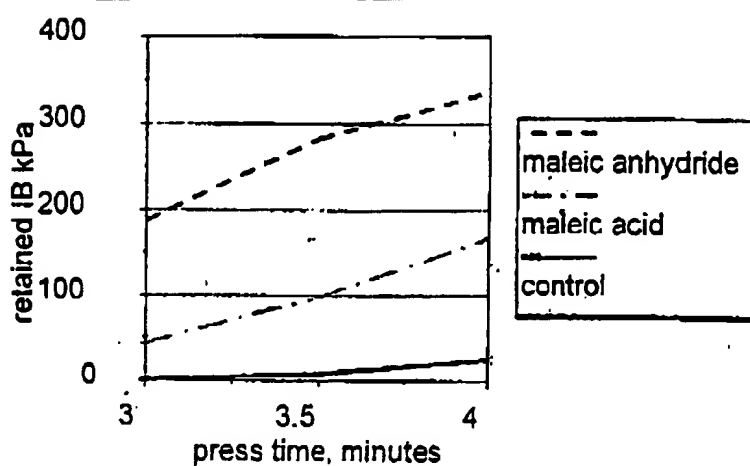


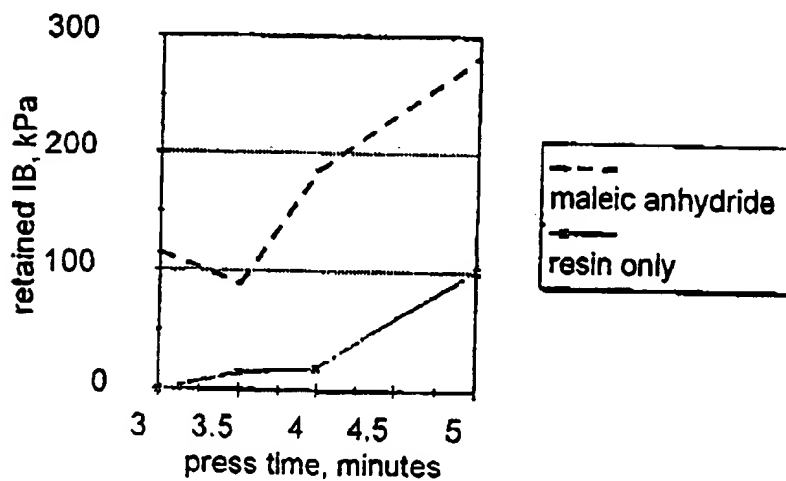
Fig.8

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Retained Internal Bond after soakingFig.9



## INTERNATIONAL SEARCH REPORT

Inte  
onal Application No  
PCT/GB 99/02420

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to/claim No.
A	WO 97/32074 A (WINDSOR TECHNOLOGIES LIMITED; SYMONS MICHAEL WINDSOR (ZA)) 4 September 1997 (1997-09-04) page 5, line 13 - line 28 page 6, line 16 - line 22 page 8, line 1 - line 10 page 10, line 14 - line 20	1
A	US 5 520 777 A (SHISKO WALTER S) 28 May 1996 (1996-05-28) cited in the application abstract; claim 1	1

Form PCT/ISA/210 (continuation of second sheet) (July 1992)

## INTERNATIONAL SEARCH REPORT

information on patent family members

Int. l. Application No

PCT/GB 99/02420

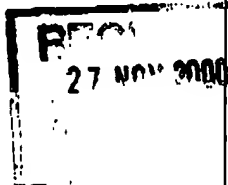
Patent document cited in search report		Publication date	Patent/family member(s)	Publication date
JP 47018996	B		NONE	
US 3968294	A	06-07-1976	US 4044087 A	23-08-1977
WO 9732074	A	04-09-1997	AU 706508 B	17-06-1999
			AU 1803797 A	16-09-1997
			EP 0880619 A	02-12-1998
US 5520777	A	28-05-1996	AU 1803195 A	11-09-1995
			CA 2184268 A	31-08-1995
			WO 9523055 A	31-08-1995

## PATENT COOPERATION TREATY

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NOTIFICATION OF TRANSMITTAL OF  
THE INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT

(PCT Rule 71.1)

Date of mailing  
(day/month/year) 23.11.2000

Applicant's or agent's file reference  
PBA/DO88449PWO

## IMPORTANT NOTIFICATION

International application No.  
PCT/GB99/02420

International filing date (day/month/year)  
06/08/1999

Priority date (day/month/year)  
06/08/1998

Applicant

UNIVERSITY OF WALES, BANGOR et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

## 4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

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## PATENT COOPERATION TREATY

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## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference <b>PBA/DO88449PWO</b>		<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. <b>PCT/GB99/02420</b>	International filing date (day/month/year) <b>06/08/1999</b>	Priority date (day/month/year) <b>06/08/1998</b>	
International Patent Classification (IPC) or national classification and IPC <b>B27N3/00</b>			
Applicant <b>UNIVERSITY OF WALES, BANGOR et al.</b>			
<p>1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 5 sheets, including this cover sheet.</p> <p><input type="checkbox"/> This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).</p> <p>These annexes consist of a total of sheets.</p>			
<p>3. This report contains indications relating to the following items:</p> <ul style="list-style-type: none"> <li>I <input checked="" type="checkbox"/> Basis of the report</li> <li>II <input type="checkbox"/> Priority</li> <li>III <input type="checkbox"/> Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</li> <li>IV <input type="checkbox"/> Lack of unity of invention</li> <li>V <input checked="" type="checkbox"/> Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability, citations and explanations supporting such statement</li> <li>VI <input type="checkbox"/> Certain documents cited</li> <li>VII <input type="checkbox"/> Certain defects in the international application</li> <li>VIII <input checked="" type="checkbox"/> Certain observations on the international application</li> </ul>			
Date of submission of the demand <b>03/03/2000</b>		Date of completion of this report <b>23.11.2000</b>	
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465		Authorized officer  <b>Baath, C</b>  Telephone No. +49 89 2399 2959	

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/GB99/02420

**I. Basis of the report**

1. This report has been drawn on the basis of *(substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments (Rules 70.16 and 70.17).)*  
**Description, pages:**

1-12 as originally filed

**Claims, No.:**

1-24 as originally filed

**Drawings, sheets:**

1/9-9/9 as originally filed

2. With regard to the language, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**International application No. **PCT/GB99/02420**☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement****1. Statement**

Novelty (N)	Yes:	Claims	
	No:	Claims	1-3
Inventive step (IS)	Yes:	Claims	4, 6-17, 21-24
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	1-24
	No:	Claims	

2. Citations and explanations  
see separate sheet

**VIII. Certain observations on the international application**

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:  
see separate sheet

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P.37/42

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/GB99/02420

**VIII Clarity**

1. Current claim 5 contradicts the teaching of current claim 4. Although disclosed in the current application, the conditions under which such a mixing with the phenol resin can be tolerated without the cited adverse effect of increasing viscosity has not been discussed. The requirements of Art. 6 and Rule 5.1 iii) are not satisfied. Moreover, mixing the maleic agent with the phenol formaldehyde agent is known from the said JP abstract, Art. 33 (2) is not met.
2. Claim 18 appears to be a mere repetition of what is claimed in claim 1. Claim 18 contradicts the requirement of conciseness, Art. 6. The same applies to claims 19 and 20.